



# Carbon additives for electrical double layer capacitor electrodes



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## HIGHLIGHTS

- Rate capability is clearly enhanced using carbon additives.
- The ESR and EDR can be reduced by up to 30%.
- 5 wt.% carbon additive does not significantly affect the specific capacitance.
- Electrode processing can be affected by the additives.

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## ABSTRACT

Electrochemical double layer capacitors (EDLCs) are inherently high power devices when compared to rechargeable batteries. While capacitance and energy storage ability are mainly increased by optimizing the electrode active material or the electrolyte, the power capability could be improved by including conductive additives in the electrode formulations. This publication deals with the use of four different carbon additives – two carbon blacks and two graphites – in standard activated carbon based EDLC electrodes. The investigations include: (i) physical characterization of carbon powder mixtures such as surface area, press density, and electrical resistivity measurements, and (ii), electrochemical characterization via impedance spectroscopy and cyclic voltammetry of full cells made with electrodes containing 5 wt.% of carbon additive and compared to cells made with pure activated carbon electrodes in organic electrolyte. Improved cell performance was observed in both impedance and cyclic voltammetry responses. The results are discussed considering the main characteristics of the different carbon additives, and important considerations about electrode structure and processability are drawn.

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## 1. Introduction

Electrodes for electrochemical double layer capacitors (EDLCs) are typically prepared from activated carbon (AC), which in most cases is bound to electrode sheets using a polymeric binder [1–3]. Contrary to Li-ion batteries, carbon additives are not commonly used in EDLCs. The use of carbon additives such as graphites and carbon blacks is a state-of-the-art method to improve the conductivity of cathodes in Li-ion batteries, often based on low conductive active materials [4]. AC used in EDLCs is intrinsically much more conductive than typical battery cathode materials. In spite of this, in many publications conductive carbons are added to AC, often without a discussion of the beneficial or even detrimental

effects of the additives. Pandolfo et al. [5] investigated the effects of the addition of carbon black and graphite to AC by using electrochemical impedance spectroscopy in an organic electrolyte. These authors found that a few percent of carbon black is enough to enhance the performance of the electrode by increasing the electrical connectivity of the electrode, but also by modifying its porous structure. Large concentrations of carbon black were found to decrease the capacitance per unit mass of electrode, due mainly to the lower specific surface area of the additive.

Using aqueous electrolyte, Zhang et al. [6] investigated the effect of acetylene black on the performance of EDLC electrodes. An addition of 5 wt.% of acetylene black was found to be the best choice for the improvement of energy and power density of the system. Higher contents of acetylene black hindered ion transport by blocking diffusion pathways inside the electrode.

Jost et al. [7] used 10 wt.% of onion like carbon to enhance the conductivity of their electrode material. They could show that this addition led to a pronounced decrease in equivalent series resistance (ESR).

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Typically, the specific surface area of carbon additives is lower compared to the one of AC, which lowers the overall gravimetric capacitance and, therefore, the specific energy of the system. Considering their widespread use as carbon additives in batteries, carbon blacks and graphites are the materials of choice in this study.

Carbon blacks are generally formed by the incomplete combustion of hydrocarbons [8]. A specific category of carbon blacks is the one of conductive carbon blacks, which consist of primary particles (typically with diameters of several tens of nanometers) which are fused together to form aggregates of a complex, mostly branched, structure. The extent of the carbon network formed by these aggregates in a composite with a low conducting powder determines the electrical conductivity. As soon as the concentration of the carbon black is high enough to reach the percolation threshold (typically a few wt.%), a fully developed conductive network is established which embeds the active material particles. The optimal conductivity is achieved if, at carbon concentrations beyond the percolation threshold, these agglomerates are fully dispersed in the electrode slurry and recombine to form a conductive network of carbon black aggregates in the final electrode during the wet coating and drying process [4].

Conductive carbon blacks, due to their structure, typically have a large void volume and low apparent density, which explains a high absorption of solvent when compared to other materials like activated carbons or flake graphites.

The intrinsic conductivity of graphite is higher than that of carbon black due to the higher crystallinity. The average particle size of typical graphite powders used as conductive additive is in the range of 2–10  $\mu\text{m}$ . Because of the different morphology and higher true density, percolation takes place at a higher loading compared to conductive carbon blacks [4]. Highly crystalline graphite powders can be easily compressed, therefore they act as compaction aid when incorporated in an electrode [4].

Expanded graphite consists of particles with high aspect ratio; the particle flakes are typically much thinner than those of a flake graphite. This highly anisometric particle morphology leads to a lower percolation threshold of expanded graphite compared to flake graphite.

The incorporation of the carbon additive into AC electrodes might have an effect on the surface area and press density of the electrode and thus on the maximum volumetric and gravimetric capacitance, whereas the electrical resistivity, which is expected to be mostly affected by the carbon additive, has to be as low as possible for high power applications. In this respect the effect of single wall carbon nanotubes as carbon additive was investigated by Pico et al. [9]. It was found that the specific capacitance of the electrode is reduced according to the reduction of the surface area with increasing amount of additive and that the resistivity of the electrodes agrees with their percolation model. The amount of solvent consumed during electrode preparation is an important aspect of the electrode (material) processability.

This publication deals with the addition of four different carbon additives – a high and a low surface area carbon black as well as a flake and an expanded graphite – to standard AC used in EDLC electrodes. Physical and electrochemical characterizations are reported. Surface area, pressed density, electrical resistivity, and solvent uptake during slurry preparation was screened by studying powder mixtures of AC and carbon additives. Capacitor electrodes containing 5 wt.% of carbon additive are prepared and electrochemically characterized in full cells via electrochemical impedance spectroscopy (EIS) and cyclic voltammetry (CV) in organic electrolyte 1 M TEABF<sub>4</sub>/AN. Electrodes without any carbon additive are measured for comparison.

## 2. Experimental

### 2.1. Materials

Activated carbon YP-50F (Kuraray Chemical, Japan, hereafter called AC) is commercially available and was used as received.

The carbon additives can be divided in two groups:

- (I) Carbon blacks: C-ENERGY™ Super C45 (Super C45) (TIMCAL SA, Bodio, Switzerland) and ENSACO 350 (TIMCAL SA, Bodio, Switzerland).
- (II) Graphites: C-ENERGY™ KS6L (KS6L) (TIMCAL SA, Bodio, Switzerland) and an Experimental Expanded Graphite (EXP) (TIMCAL SA, Bodio, Switzerland).

The electrolyte used for the electrochemical characterization was 1 M tetraethylammonium tetrafluoroborate in acetonitrile (Honeywell Specialty Chemicals Seelze, Germany, 1 M TEABF<sub>4</sub>/AN). Karl Fischer titration (684 KF Coulometer, Metrohm, Switzerland) showed a water content of below 20 ppm.

### 2.2. Powder characterization

#### 2.2.1. BET specific surface area

The specific surface area (SSA) was measured according to the Brunauer–Emmett–Teller (BET) method using an ASAP 2020 instrument (Micromeritics, USA) in the pressure range 0.04–0.26 relative pressure, unless otherwise specified. Samples were degassed at 350 °C for 12 h prior to the measurement.

#### 2.2.2. Preparation of the powder mixtures and solvent uptake

Homogeneous AC – carbon additive mixtures containing different amounts of additive (2.5, 5 and 10 wt.%) were produced by dispersing the two carbon powders, 10 g in total, in a solvent based on a 1:1 mixture by weight of 2-propanol (Sigma–Aldrich, Germany) and water (resistivity > 18 M $\Omega$  cm, Millipore, USA).

The carbon additive was pre-dispersed in the least amount of solvent for 1 min using a rotor-stator T 25 Ultra-Turrax® high-performance disperser (IKA®, Germany), equipped with an S 25 N 18 G dispersing element operated at 11'000 rpm (this first step was skipped in the case of pure AC dispersion). Then AC was added and the viscosity was empirically adjusted to get an easily flowing slurry by adding solvent. The slurry was homogenized at high shear (1 min at 11,000 rpm). The solvent was removed by drying the dispersions overnight at 80 °C. The amount of solvent used to make the dispersion is henceforth called “solvent uptake”, and it is expressed as wt.% of solvent present in the dispersion.

#### 2.2.3. Press density and electrical resistivity under compression

Pure carbon powders were used as received. The powder mixtures were prepared as reported in Section 2.2.2. A sample (0.6 g) is compressed in a non-conductive die between two electrified pistons made of brass. Compaction is achieved with an electro-mechanic 4-column uniaxial press (type PWV 4 ES-SERVO, Paul-Otto Weber Maschinen-und Apparatebau GmbH, Germany). A given current, generated by a current source (model 6221, Keithley, USA), is forced through the powder sample via the two pistons. The voltage drop across the pistons is measured with a nanovoltmeter (model 2182A, Keithley, USA). The relative position of the pistons in the die (i.e. the height of the powder sample) is measured with a length gauge (model ST 3000, Heidenhain, Germany). The two-point electrical resistance is then calculated at any given pressure from the current and voltage drop according to Ohm's law. It is assumed that the contact resistances between pistons and carbon samples are the same regardless the composition of the sample

and, hence, can be neglected for comparative purposes. Since the height, thus volume, and mass of the powder sample are known, the density and electrical resistivity can be calculated at any given pressure. Density and resistivity values reported here were measured at  $4.5 \text{ kN cm}^{-2}$ , the pressure at which AC powder has comparable density to an AC electrode prepared as described in Paragraph 2.3.

#### 2.2.4. Oil absorption number

The oil absorption number test is a means to determine the general behavior of powder materials with respect to their absorption properties. The oil absorption number (OAN) of the carbon additive is expected to have a correlation to the amount of solvent needed to prepare powder dispersion. A carbon with a higher OAN requires more solvent to be dispersed. OAN is measured using paraffin oil and an absorptometer according to the norm ASTM D2414 [10]. Briefly, oil is dosed in a mixing chamber containing the test powders. The torque of the mixing elements is measured. The torque rises steeply when the powders are fully impregnated and this determines the end of the oil dosing and consequently the OAN value.

#### 2.2.5. Particle size distribution

Particle size distribution is measured by laser diffraction with a Malvern Mastersizer S system. The 90th percentile (D90) of the distribution is reported.

#### 2.3. Electrode preparation

Self-standing electrode sheets were prepared from a slurry containing 90 wt.% of carbon material and 10 wt.% of polytetrafluoroethylene binder (PTFE, 60 wt.% dispersion in  $\text{H}_2\text{O}$ , Sigma–Aldrich). When carbon additives are used, the carbon material consists of 95 wt.% AC and 5 wt.% carbon additive.

The carbon material was dispersed in a 1:1 mixture of isopropanol:water and ultrasonicated for 15 min before the slurry was heated and vigorously stirred until a dough like mass was achieved. This mass was subsequently rolled into self-standing electrode sheets with a thickness of about  $206 \pm 15 \mu\text{m}$ . After drying for at least 24 h at  $120^\circ\text{C}$  and 20 mbar, a piece of  $5 \times 5 \text{ cm}$  was cut out from the sheets, put between two stainless steel plates and compressed for 10 min with a force of  $10 \text{ t}$  ( $400 \text{ kg cm}^{-2}$ ). This led to a reduction of thickness of about 10 %. Afterward, discs with a diameter of 12 mm (loading: approx.  $13 \text{ mg cm}^{-2}$ ) were punched out of the compressed sheets and used for electrochemical characterization.

#### 2.4. Electrochemical characterization

For electrochemical characterization a homemade electrochemical cell was used. The electrodes were sandwiched between

two spring loaded ( $50 \text{ N cm}^{-2}$ ) Ti pistons. Two carbon coated aluminum current collectors (z-flo 2653, Exopack Advanced Coatings, USA) were used for better contact. The two electrodes were separated using a  $30 \mu\text{m}$  thick paper separator, resulting in a symmetric capacitor configuration. Before the cell was transferred to the glove box for electrolyte filling, it was again dried at  $120^\circ\text{C}$  and 20 mbar for at least 8 h.

First cyclic voltammetry (CV) was performed at  $10 \text{ mV s}^{-1}$  in the range of  $\pm 500 \text{ mV}$  cell voltage for 3 cycles to ensure electrode wetting and as a first performance check. Afterward, the cell was characterized by means of electrochemical impedance spectroscopy (EIS). The cell voltage was kept at 0 V for 10 min before the impedance spectrum was recorded at an amplitude of 5 mV and in a frequency range of 10 MHz–100 kHz. Subsequently the rate capability was tested using CV at different scan rates (1, 2, 5, 10, 50, and  $100 \text{ mV s}^{-1}$ , only tests performed at 1 and  $100 \text{ mV s}^{-1}$  are commented in the text) in the voltage range from 0 to 2.5 V. Each scan rate was applied during 3 cycles. All electrochemical measurements were performed at room temperature using a VMP3 system (Bio-Logic, France).

The reported gravimetric values refer to the total carbon contained in the cell, therefore AC and the carbon additive, or only AC in the case of AC electrode without carbon additive.

### 3. Results and discussion

#### 3.1. Powder characterization

Some properties of the used carbon additives are listed in Table 1. Clearly, the SSA of the carbon additives is much smaller than the one of the AC. The decrease in SSA upon addition of 5 wt.% carbon additive is roughly proportional to the difference in SSA between AC and carbon additive. Consequently, high surface area carbon additives are expected to lead to a smaller reduction in gravimetric capacitance.

##### 3.1.1. Press density

The press density of the powder mixtures was determined as described in Section 2.2.3. The press densities as a function of carbon additive amount are shown in Fig. 1a. For carbon additive amounts above 2.5 wt.%, an increase in press density is observed for all additives. The trend is more pronounced for the graphite additives, which are compacted more easily compared to carbon blacks due to their compressibility. In addition graphite likely reduces the internal friction during pressing leading to a better compaction. At a loading of 5 wt.%, the graphite additives allow for a significant density increase. The press density of the powder mixtures containing carbon black also increased with high loading. Most probably the small carbon black particles fill the voids between the AC particles, as suggested by Pandolfo et al. [5].

**Table 1**  
Relevant properties of the used carbon additives and AC.

Material name	Material type	BET SSA	BET SSA of the 5 wt.% mixture in AC <sup>a</sup>	D90	OAN	Electrical resistivity
		$\text{m}^2 \text{g}^{-1}$	$\text{m}^2 \text{g}^{-1}$	$\mu\text{m}$	$\text{ml}/100 \text{ g}$	$\Omega \text{ cm}$
Super C45	Low surface area carbon black	45	1461	— <sup>c</sup>	335	0.044
ENSACO 350	High surface area carbon black	770	1544	— <sup>c</sup>	351	0.044
KS6L	Flake graphite	20	1483	6	113	0.027
EXP	Expanded graphite	27	1461	40	187	0.026
AC	Activated carbon (active material)	1561 <sup>a</sup>	— <sup>b</sup>	10	167	0.138

<sup>a</sup> Single point BET at 0.18 relative pressure.

<sup>b</sup> Not applicable.

<sup>c</sup> Not applicable. The determination of PSD via regular laser techniques is meaningless for conductive carbon blacks due to their branched structure.

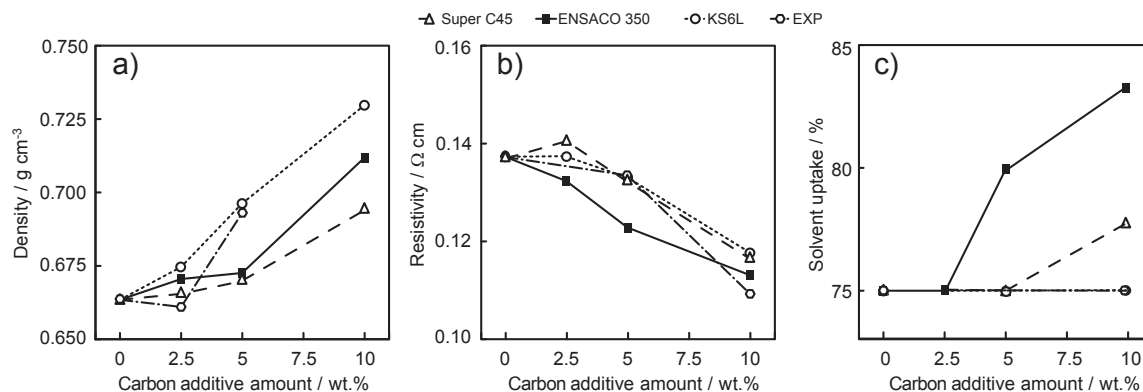


Fig. 1. (a) Press density, (b) electrical resistivity, and (c) solvent uptake of the powder mixtures as a function of carbon additive amount.

### 3.1.2. Electrical resistivity

The electrical resistivities of the various pure materials are reported in Table 1. Clearly the carbon conductive additives have a significantly lower resistivity compared to activated carbon. As expected the resistivity of the pure graphite additive is lower than the resistivity of carbon blacks. Although it has to be kept in mind that the resistivity of an electrode or mixture is primarily determined by the percolation of the additive in the matrix.

The results of the electrical resistivity measurements on powder mixtures are plotted in Fig. 1b. Similar to the effect of the carbon additives on press density, the electrical resistivity of the mixtures is decreasing for loadings higher than 2.5 wt.%. The electrical resistivity decrease is most pronounced for the carbon black ENSACO 350; a 5 wt.% loading of this high surface area carbon black leads to a significant electrical resistivity decrease.

### 3.1.3. Solvent uptake

The solvent uptake is a very important factor for industrial electrode manufacturing. An excessive amount of solvent used during electrode preparation might lead to a more expensive drying procedure since the drying might require longer time and/or more energy to remove the solvent. In addition, the drying of low solid content dispersion might lead to shrinkage and cracking of the final electrode coating. The values of solvent uptake observed during the preparation of the powder mixtures and pure AC are plotted in Fig. 1c. Loadings higher than 2.5 wt.% of ENSACO 350, or

higher than 5 wt.% of Super C45, lead to an increase of solvent uptake. The increase in solvent uptake correlates with the large OAN of carbon black compared to graphites (compare Table 1), which do not lead to an increase in solvent uptake.

## 3.2. Electrochemical performance

The powder characterization indicated that at least 2.5 wt.% of carbon additive is required to improve conductivity and press density. In addition, no more than 6 wt.% of SSA was lost upon addition of 5 wt.% additive. Therefore, all electrochemical measurements were performed on electrodes containing either 0 wt.% or 5 wt.% of carbon additive.

### 3.2.1. Electrochemical impedance spectroscopy

The impedance spectra of all tested cells are depicted in Fig. 2a as Nyquist plots. All EIS spectra are characterized by a 45° region followed by an almost vertical line representing the capacitive part of the spectrum. The value of the real impedance at 1 kHz, which corresponds to the intersection of the 45° region with the x-axis, was taken as the equivalent series resistance (ESR).

The equivalent distributed resistance (EDR), corresponding to the 45° region, is characteristic of the pore structure of the system, given the fact that the thickness of all electrodes was the same. The EDR was determined by extrapolating the 90° region to the x-axis

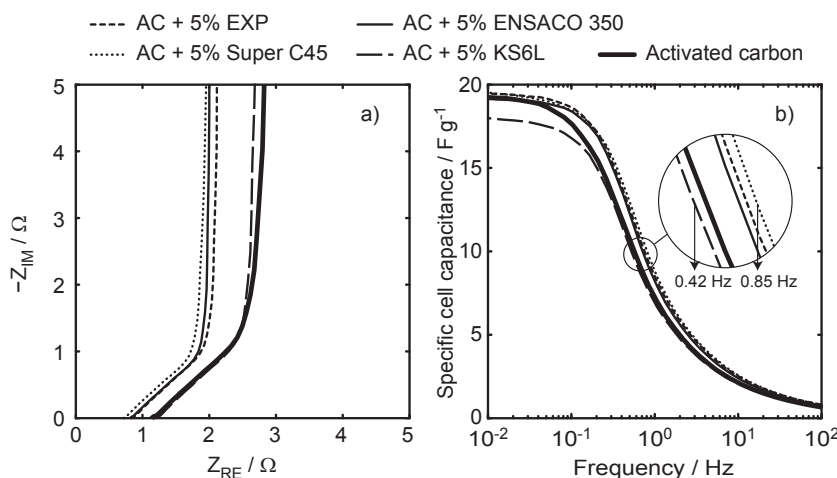


Fig. 2. (a) Nyquist plot for the systems as indicated. (b) Specific cell capacitance as a function of the frequency for the systems as indicated. Inset: magnification at half-capacitance area.

**Table 2**

ESR and EDR values for the cells based on the various electrode materials as indicated.

Electrode material	ESR	EDR
	$\Omega$	$\Omega$
ENSACO 350 (5 wt.% in AC)	0.8	1.0
Super C45 (5 wt.% in AC)	0.8	1.0
KS6L (5 wt.% in AC)	1.1	1.4
EXP (5 wt.% in AC)	0.9	1.0
AC	1.2	1.3

and subtracting the ESR from the intersection, as described in Ref. [1]. The ESR and EDR values are presented in Table 2.

A dependence on the investigated carbon mixture is visible for both ESR and EDR. All additives decrease the ESR. The effect for the addition of carbon blacks and EXP is most pronounced. The flake graphite KS6L only shows a minor impact on the ESR. This reduction is likely due to the increased electrical conductivity through the electrode material via the conductive network of the carbon blacks and the expanded graphite. Due to the lower aspect ratio of the flake graphite particles in comparison to EXP particles, it can be assumed that the conductive network is not fully developed at the given concentration of KS6L. For the EDR region, the value for the electrodes containing flake graphite is even slightly increased as compared to the bulk AC material. For EXP and the carbon blacks, the value of the EDR is decreased by more than 20%, resulting in higher pore conductivity and an overall performance increase of the system. The improved conductive network would lead to a better material utilization and therefore to a greater porosity accessible at a given frequency.

Fig. 2b shows the impedance data plotted as cell capacitance vs. frequency. As indicated in the plot, a shift to the right represents a faster response of the system, which is the case for the addition of the two carbon blacks and expanded graphite, as already seen in the ESR and EDR values. The cell capacitance value at low frequencies (upper left corner in Fig. 2b) shows that a clearly measurable cell capacitance loss is observed only for flake graphite, whereas for the other three additives virtually no cell capacitance loss is visible as compared to the electrode prepared from bare AC. The loss of surface area due to the addition of the carbon additives

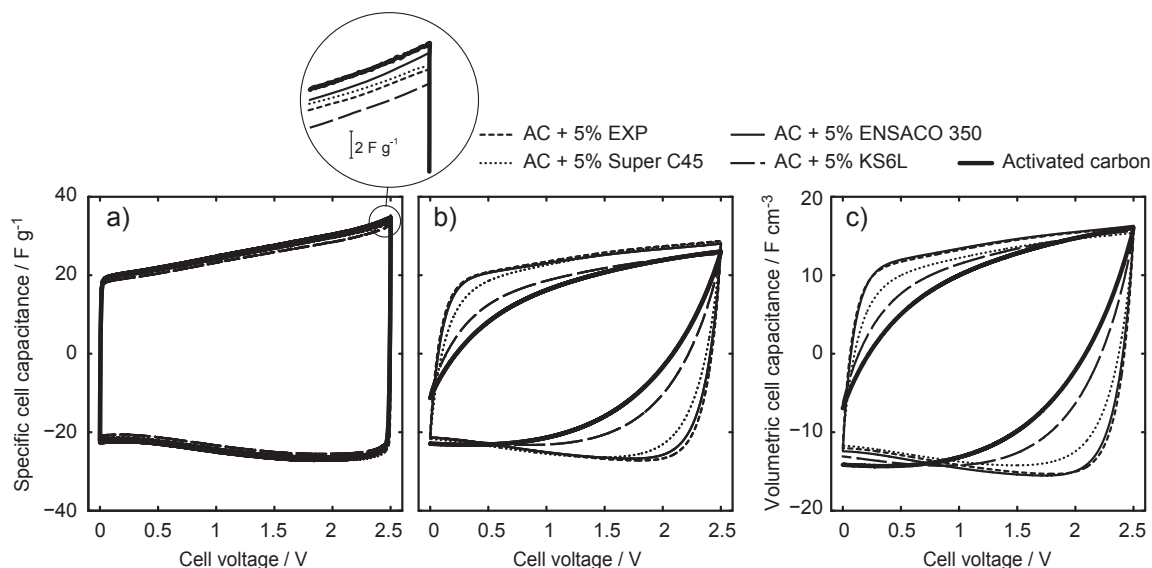
is compensated by the higher electrode conductivity, which likely translates to better connectivity in the electrode and the better material utilization.

### 3.2.2. Cyclic voltammetry

As another electrochemical measurement method, cyclic voltammetry (CV) at different scan rates was performed to either determine the rate performance of the system. After the EIS measurements, the cells were cycled up to 2.5 V cell voltage starting with a scan rate of  $1 \text{ mV s}^{-1}$  (shown in Fig. 3a). A nice rectangular CV shape is observed for all electrode compositions. At this scan rate, the rate capability does not seem to be affected by the additives. However, the specific cell capacitance is slightly reduced, as it is shown in the inset of Fig. 3a, most likely due to the overall reduced SSA. At this scan rate, the bare AC electrodes show the highest specific cell capacitance, the largest decrease is found for the addition of flake graphite KS6L. The trend in specific cell capacitance measured with CV nicely follows the trend measured with EIS and the decrease of the SSA values.

Upon increasing the scan rate, the beneficial effects of the carbon additives become clearly visible. Fig. 3b shows the specific cell capacitance of the same cells cycled at a scan rate of  $100 \text{ mV s}^{-1}$ . The cells containing bare AC and flake graphite as carbon additive suffer from a pronounced cell capacitance loss at the vertex potential (see Fig. 3b at 2.5 V) and a distortion from the rectangular shape. The cells containing carbon black or EXP show a nice rectangular shaped CV and a fast charge reversal at the vertex potentials. While the bare AC electrodes perform best in the case of  $1 \text{ mV s}^{-1}$ , at a scan rate of  $100 \text{ mV s}^{-1}$  the electrodes containing EXP outperform all other compositions. The better electrical conductivity as well as an improved material utilization and connectivity might explain the improved rate performance observed.

Another important value which has to be considered is the volumetric cell capacitance. Electrode density not only depends on the density of powder mixtures but also on the way the electrode relaxes after compaction. The effect of the changed density is visible in Fig. 3c, where the volumetric cell capacitance is plotted vs. the cell voltage. The volumetric cell capacitance values at the vertex potential are leveling out, however still retaining the much faster charge reversal upon addition of EXP or carbon blacks.



**Fig. 3.** (a) CVs measured at  $1 \text{ mV s}^{-1}$  of the systems as indicated, (b) CVs measured at  $100 \text{ mV s}^{-1}$  of the systems as indicated, (c) CVs measured at  $100 \text{ mV s}^{-1}$  of the systems as indicated reported as volumetric cell capacitance.



The above results for cell capacitance, rate capability and solvent uptake do not indicate a clear favorite among the four tested carbon additives. EXP appears to be the best choice for rate capability and solvent uptake, while ENSACO 350 shows the best specific cell capacitance at low scan rate, close to pure AC. In order to pick the right additive the different properties have to be assessed carefully.

It has to be kept in mind that the presented results apply exclusively to the investigated material combinations. For a different activated carbon or a different electrolyte the results may differ.

#### 4. Conclusions

With this publication we show the different and sometimes complementary effects of various types of carbon additives on the performance of activated carbon electrochemical double layer capacitor electrodes, taking into consideration also processing aspects. Nevertheless this study might be further extended to consider aspects like the effect of the additive concentration and synergies between different types of additives. Based on the presented work the following conclusions can be drawn:

1. Carbon conductive additives decrease the electrical resistivity of AC powder mixtures.
2. The ESR and the EDR of the cell are influenced by the addition of carbon additives. The addition of conductive carbon black or expanded graphite results in an ESR reduction of ca. 30% (at 5 wt.% additive addition). The reduction of the ESR and EDR is presumably due to the formation of a conductive network, leading to a better utilization of the active material at higher rates.
3. Specific gravimetric and volumetric cell capacitances at low rate are not significantly affected by the addition of 5 wt. % of carbon additive.
4. Conductive additives have a clear effect on the rate capability of electrodes. For the rate capability of the AC electrodes and the AC electrodes containing carbon additives, a clear trend can be

observed for the conductive carbon types: AC ~ flake graphite KS6L < low surface area carbon black Super C45 ~ high surface area carbon black ENSACO 350 ~ expanded graphite EXP, which was indicated from both CV and EIS measurements.

5. The solvent consumption is only remarkably increased in the case of carbon black addition, while graphite does not affect the processability of AC in this regard.

The overall effect of a carbon additive in an activated carbon electrode is clearly a complex combination of various effects related to both performance and manufacturing. The choice of the additive should be made carefully based on the electrode and processing requirements.

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